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Journal of Power Sources, Vol: 79, Issue: 1, pp. 30-36, May 1999

Title:
Non-conventional **fuel** cell systems: new concepts and development

Authors:
Zhu, Bin[a]; Meng, Guangyao[b]; Mellander, Bengt-Erik[c]

Affiliations:

a. Department of Chemical Engineering and Technology, Royal Institute of Technology, S-100 44 , Stockholm, Sweden
b. Department of Materials Science and Engineering, University of Science and Technology of China, 230026 , Hefei, Anhui, China
c. School of Physics and Engineering Physics, Chalmers University of Technology and Göteborg University, S-412 96 , Gothenburg, Sweden

Address:
(No address specified)

Keywords:
Fuel cells; Non-conventional; Single chamber; Single component fuel cell; Gadolinium-doped ceria; Salt–ceramic composite; Ceria-based oxides; Soft-chemistry process

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Publisher:
Elsevier Science

Language of Publication:
English

Item Identifier:
S0378-7753(98)00195-5

Publication Type:
Full Length Article

ISSN:
0378-7753

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Journal of Power Sources 79 (1999) 30–36

JOURNAL OF POWER SOURCES

Non-conventional fuel cell systems: new concepts and development

Bin Zhu [a,*], Guangyao Meng [b], Bengt-Erik Mellander [c]

[a] Department of Chemical Engineering and Technology, Royal Institute of Technology, S-100 44 Stockholm, Sweden
[b] Department of Materials Science and Engineering, University of Science and Technology of China, 230026 Hefei, Anhui, China
[c] School of Physics and Engineering Physics, Chalmers University of Technology and Göteborg University, S-412 96 Gothenburg, Sweden

Received 30 August 1998; received in revised form 18 November 1998; accepted 24 November 1998

Abstract

This paper considers fuel cell research from a new point of view, describing so called non-conventional systems including intermediate temperature and new concept fuel cells based on recent developments of materials and systems. © 1999 Elsevier Science S.A. All rights reserved.

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1. A brief overview

The ever-increasing need for new energy conversion technologies that are compatible to strict environmental requirements has made fuel cells (FCs) one of the most extensive and active research areas of today. In a FC device chemical energy (a fuel and an oxidant) is converted directly into electrical energy, without having the Carnot-limitation of the efficiency which accompany all heat engines. The FC can therefore perform the energy conversion with a considerably higher efficiency than traditional energy conversion plants. The fuel can for example be hydrogen gas, natural gas, methanol or ethanol and the oxidant be oxygen gas or air. FCs provide a very clean method of producing electrical energy, if hydrogen and oxygen gas are used as fuel and oxidant pure water is the only exhaust. However, present solid state fuel cell technology, e.g., solid oxide fuel cells (SOFC) or solid polymer fuel cells (SPFC), has met a number of problems concerning the materials and technology. This has affected the cost and led to a delayed commercialisation. New power sources are under consideration all around the world today. FCs can be used with many types of fuels and may thus provide a flexible solution also in an interim period when the society is converting from fossil fuel to renewable energy sources.

The SOFC may in some respects be seen as an ideal source to generate electricity due to its high efficiency and flexibility regarding choice of fuels. Other types of FCs often need a reformer to be able to use, e.g., natural gas as fuel. However, in current SOFC stack constructions the high temperature, ~ 1000°C, results in common problems, e.g., i) interconnecting and sealing materials do not perform properly, serious cracking, leakage and finally destruction of the stack occurs; ii) chemical stability of the FC components are more critical at high temperature [1,2]. These problems are still the major challenge for the current SOFC technology commercialisation after more than 30 years of progress.

Non-conventional systems, e.g., intermediate temperature FCs, however, provide new routes to avoid the problems associated with the high temperature SOFC plants. During the last few years the interest for intermediate temperature FCs has grown rapidly [3–15], and a considerable progress has been achieved regarding non-conventional fuel cells. It is the intention for this paper to consider the problems and future development from a new angle based on recent technological achievements.

2. New concept fuel cells

The conventional fuel cell device essentially consists of two porous electrodes separated by a dense proton (or oxygen) conducting electrolyte. The construction includes

* Corresponding author. Tel.: +46-8-7908419; Fax: +46-8-108579; E-mail: binzhu@ket.kth.se

PII: S0378-7753(98)00195-5

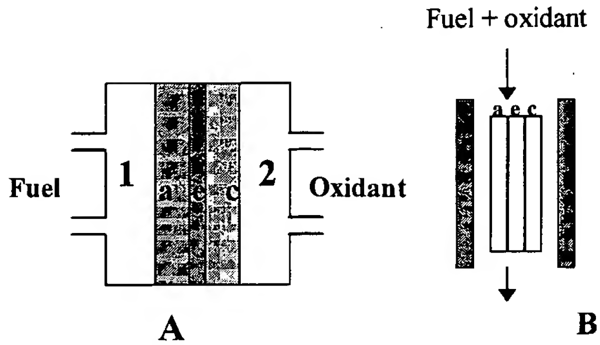

Fig. 1. Various fuel cell constructions. A) Conventional fuel cell, a: anode, e: electrolyte, c: cathode, 1: fuel chamber, 2: oxidant chamber; B) a schematic example of a single chamber fuel cell.

two chambers to supply oxidant and fuel to the electrodes, see Fig. 1a. In this article, we will mainly discuss this type of FCs where new materials have been used for electrolyte and electrodes.

Two new concepts for FCs will also be mentioned. Single chamber FCs have been proposed, e.g., by Dyer [16], and a single chamber FC has been developed as a practical device lately [17,18]. This type of FC uses a mixture of oxidant and fuel in a single chamber in order to solve the sealing problem of conventional FCs, and provide a simple system and technology. Fig. 1b shows a schematic of the principle of this device according to Ref. [17].

Another new concept is the single component device [19], where an ionic conductor is doped in order to obtain an electronic conducting electrode at each side. This type of FC, which in principle may be used either in the two- or single chamber configuration, is expected to solve interfacial and incompatibility problems among the cell components.

Based on the FC technology, many cogeneration systems may be constructed. The single chamber FC mentioned above [17], in fact, is a cogeneration system, which offers an energy conversion and a cogeneration of useful chemicals at the same time. Another example that uses a two chamber FC is the desulfurization/FC cogeneration system that will be described later. This cogeneration offers a solution to two important issues: removing an industrial pollutant and simultaneously producing electricity in an efficient way.

3. Materials and technical developments

The development of materials for non-conventional fuel cells is here mainly attributed to alternative electrolytes for the intermediate temperature region, both regarding oxygen ion and proton conductors. Soft chemical synthesis techniques can be used for material preparation, such as polymeric precursors and chemical co-precipitation sol–gel, and thin or thick films may be produced using silk screen printing, doctor-blade technique, CVD and MOCVD etc. Both porous and dense membranes such as α-Al_2O_3, γ-Al_2O_3, AlN, CeO_2, ZrO_2, TiO_2, YSZ, Li_2SO_4-based composite materials, as well as metals or alloys (e.g., Pd, Pd–Ni, Pd–Y) etc., have been extensively investigated by us. For example, a novel extrusion technique for tubular porous ceramic supports (pore size 1 to 10 μm) and a modified suspension method for microfiltration membranes (pore size 0.1 to 1 μm) on α-Al_2O_3 supports have been developed at USTC (University of Science and Technology of China) [20–22]. A number of other soft chemical techniques for both uniform porous or dense membranes, such as sol–gel [23,24], plasma CVD [25] and MOCVD [26] have recently been developed at USTC.

3.1. Oxygen ion conducting electrolytes—bulk and thin film ceramic membranes

For oxygen ion conducting ceramics, intensive work has been focused on improving gadolinium-doped ceria $Ce_{1-x}Ce_xO_{2-y}$ (GCO) and $La_{0.8}Sr_{0.2}Ga_{0.8}Mg_{0.8}O_3$ (LSG-M). Doping plays a key role in tailoring the electrical conductivity and other properties in these oxides. By doping, the electrical properties can be dramatically varied between ionic and electronic conduction, which gives a flexibility to obtain the required function for the electrolyte or the electrode. Nanostructured oxides can have an enhanced oxygen vacancy mobility and redox reactivity. High catalyst function, sulphur tolerance, high conductivity, and chemical stability make these materials attracting and functional for intermediate temperature FC applications.

It should be pointed out that although CeO_2-based fluorite type oxides are often used in intermediate temperature SOFCs, there is a critical issue concerning chemical stability and electronic conduction. It has been reported that YSZ and Sm-doped ceria (SDC) composite electrolytes can effectively suppress the electronic conduction of the ceria-based electrolytes [27]. In this composite electrolyte, SDC grains were dispersed in a YSZ matrix, so that the electronic conduction of the SDC was effectively blocked by the YSZ matrix. A SOFC with this composite electrolyte offered the advantages of a higher cell voltage due to pure ionic conducting YSZ, and high conductivity from SDC. The SDC content and the particle size significantly affected the FC power generation characteristics, and the polarisation was significantly decreased. In parallel, our recent efforts focus on i) developing various nanostructured doped ceria-based thin films, which are highly conductive and stable in the fuel atmospheres, e.g., hydrogen [28,29]. Using these films coated on GCO electrolytes may be expected to give a better compatibility, stability and higher exchange current than YSZ/GCO layers, which are used to protect the GCO electrolyte from

reduction and prevent electronic conduction [30,31]; ii) composite ceria electrolytes using second phase materials, e.g., salts and oxides, in order to suppress efficiently electronic conduction and significantly enhance the FC efficiency [32] in a similar way to that of the SDC–YSZ composite.

3.2. Proton conducting materials— bulk and film ceramic membranes

Salt-oxide ceramic composites (SOCs), e.g., Li_2SO_4 (or other salts)–Al_2O_3 (or $-SiO_2$) composites may be very functional with good stability in strongly reducing atmospheres. These materials have been successfully demonstrated for intermediate temperature FC applications [4–6]. A new break-through has been achieved by preparing the SOCs containing a molten phase, e.g., NaOH–NaCl–Al_2O_3, using a small amount of the molten salt mixture incorporated into the interfacial regions of the host oxide grains with controlled microstructure. In this case a very small amount of the molten salt can cause a high conductivity due to a so called composite effect [33–36], but not weaken the mechanical strength. Thus the high ionic conduction $> 10^{-2}$ S/cm in the intermediate temperature region, or other desired property, of the molten phase, as well as the chemical stability and mechanical strength of the oxide can benefit the system at the same time.

Very recently, proton conduction in chlorides and fluorides as well as their composites have been reported. These materials have also been suggested for intermediate temperature FC applications [37,38]. Fig. 2 shows some typical I–V characteristics of FCs using MF_x based electrolytes and $LiMO_2$ (M = Ni and Co) with a rock salt structure as electrodes. As can be seen in Fig. 2, a current density of 200 mA/cm^2 has been achieved near 0.4 and 0.6 V respectively for the BaF_2–Al_2O_3 and NaF–CaF_2–Al_2O_3 electrolyte FCs at 750°C. A peak power density of 0.11 W/cm^2 at 250 mA cm^{-2} has been achieved for the NaF–CaF_2–Al_2O_3 electrolyte FC. These results have been demonstrated based on bulk materials without any technical efforts for practical devices, e.g., using ceramic membrane technology to prepare dense electrolytes. Further studies are thus being carried out using tape casting technique to scale up the planar type FC to the large area needed for the practical FC devices/stack.

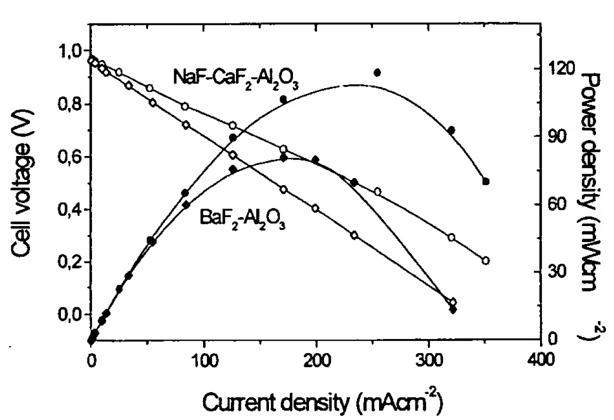

Fig. 2. Some typical I–V characteristics from fluoride electrolyte fuel cells at 750°C.

In the material development, nanometer materials are an important issue for achieving high performance FCs. Using thin film technology, more commonly via a soft-chemical route, e.g., sol–gel and suspension methods, the size range around or below 10 nm can be prepared in a highly cost-effective way. These nanostructed materials, often in form of thin films, can have various properties as a result of a variety of confinement effects, e.g., often mechanical or electrical properties will be altered compared to bulk materials. Sol–gel etc. soft chemical processes are one of the most economic ways of obtaining nanophase materials. Nanostructured materials and technology have become very important for preparing thin film, dense electrolytes, where a good control of the size and morphology of the phase domains is needed.

3.3. Novel desulfurization and fuel cell co-generation— new advanced environment / energy technology

H_2S is a noxious gas with an annual production rate of more than 10 million tons mainly from the oil and gas industry. The standard method for handling H_2S is to partially oxidise the hydrogen sulphide in 'Claus' reactors, i.e.,

$$2H_2S + O_2 \rightarrow 2H_2O + S_2 \tag{1}$$

to elemental sulphur and water. To use H_2S as fuel in a FC is, however, obviously more desirable and beneficial. Some efforts have been devoted to using SOFCs to covert H_2S to sulphur [39–44], but these have not been successful due to the reaction of the oxide electrolyte with H_2S. Using a proton conducting sulphate-based electrolyte a FC device may have the following reactions:

$$\text{at anode: } 2H_2S \rightarrow S_2 + 4H^+ + 4e^- \tag{2}$$

and

$$\text{at cathode: } 4H^+O_2 + 4e^- \rightarrow 2H_2O \tag{3}$$

The sulphate-based electrolytes, e.g., Li_2SO_4–Al_2O_3 previously used for FCs with H_2 as fuel [4,5,13], perform very well with H_2S as fuel because of their chemical stability with H_2S. We have succeeded to obtain high-conductive, mechanically strong and dense Li_2SO_4–Al_2O_3 film ceramic membranes on the porous alumina tubes for this purpose [14,45]. Fig. 3 shows a single tubular fuel cell device using Li_2SO_4–Al_2O_3 as the electrolyte on a porous

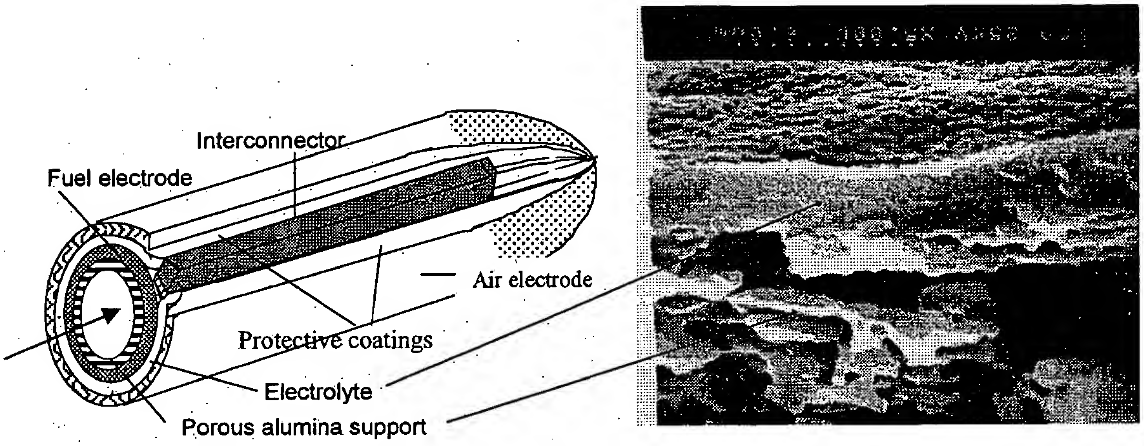

Fig. 3. Single tubular fuel cell devices where a key technology is to prepare dense film ceramic electrolyte membranes on the porous alumina tubes, see SEM photo.

alumina support. The SEM photo shows the successful preparation of a dense Li_2SO_4–Al_2O_3 ceramic film on the porous alumina tube (40% porosity), a key technology for the device construction. The preparation is performed using the sol–gel and suspension technique, which is available for large scale highly cost-effective production. For desulfurisation/FC cogeneration a technical break-through has recently been achieved by obtaining a short circuit current density > 200 mA/cm^2 and power density > 70 mW/cm^2 for the device using Li_2SO_4–Al_2O_3 materials. The device has been continuously operated with stable current output. During the operation, S_2 and water were collected from the anode and cathode, respectively, indicating the success in desulfurization and electricity co-generation. This research has thus progressed to a technical development for practical co-generation devices/plant [37].

3.4. System construction

At both the research and development stage basic choices regarding materials and designs are being made and validated; decisions must incorporate considerations regarding system cost, environmental impact, system integrability, manufacturability and marketability. Based on these research achievements the intermediate temperature FC stack is currently being investigated, and constructed in two types, tubular and planar, described separately as follows.

Fig. 4 shows a tubular intermediate temperature FC plant, where the single tubular devices (see Fig. 3) are connected in parallel and in series. The main advantage of this type of design is that highly cost-effective Al_2O_3 tubes are used in the construction, which provides possibilities for an economic fabrication in a scaled-up future industry. In addition, alumina supports have good compatibility with the electrolyte using alumina as a composite phase. The integrated single tube FC device/stack is related to this type of tubular FCs, see Fig. 5, where a number of separate cells are connected in series along a single tube. By extending the connection of these single tube stack units in parallel or in series, a powerful FC stack can be obtained.

It should be addressed that this type of single unit stack can also be constructed as a plate, similar to the construction of integrated electronic circuits. This design provides a way to construct an intermediate temperature FC stack using advanced technology from the electronic industry. In the future, a FC stack consisting of individual integrated plates installed into a framework would be possible, since the operating temperature is in the intermediate region, where normal metal and alloy materials can be used for the construction. More details about this novel integrated FC stack are contained in a newly registered Swedish patent application [46].

For the intermediate temperature FC tubular stack construction the following cell and ancillary materials are used: support: porous Al_2O_3 tubes; anode: nickel oxide (doped or composites); electrolyte: CeO_2-based oxides (doped or composites) or Li_2SO_4–Al_2O_3 (–SiO_2) composites; cathode: LaSrCoFeO; or $LiMO_2$ (M = Ni, Co); interconnect: nickel–aluminium alloy, or stainless steel. In order to ensure a good function of the FC components, multi-layer-structured films are employed. For the tubular type FCs, the soft-chemical route is favoured for fabrication due to the special geometry, also because of its unique advantages, e.g., the flexibility in selection of materials, easy preparation, the possibility to produce large sizes and complicated shapes, low cost and widely available technology for the industry.

A new design based on single chamber FCs is proposed for the future intermediate temperature FC technology, see Fig. 6, where the FC stack fed by a fuel (natural gas)/air mixture. In this construction the electrolyte film is coated

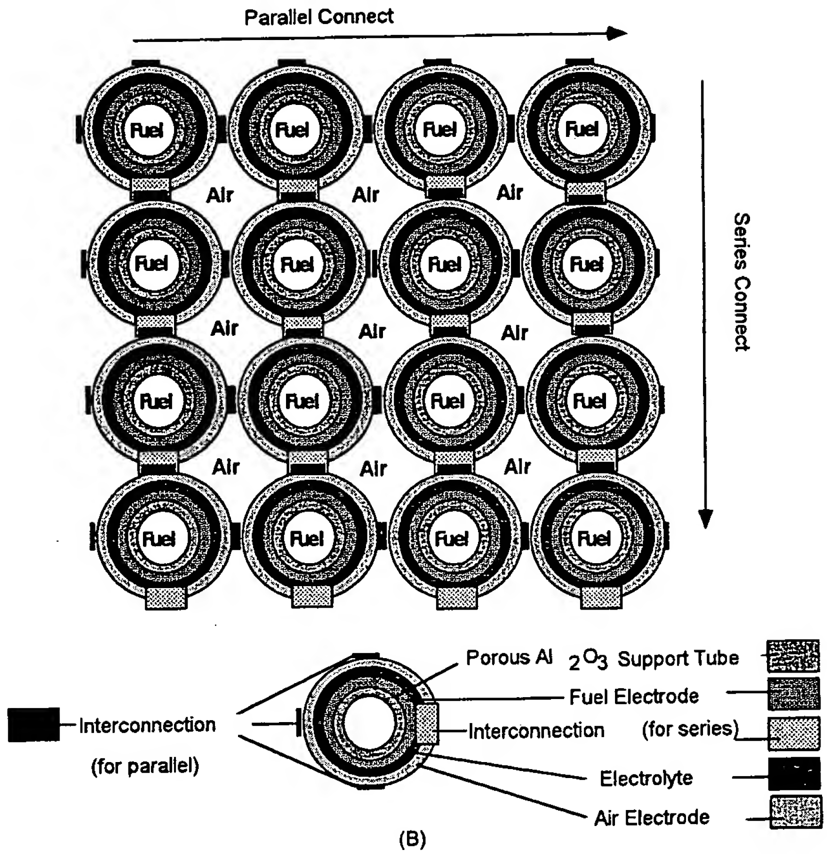

Fig. 4. Tubular FC plant constructed by connecting single devices in parallel and in series.

on both sides of an alumina support sheet and the electrodes as well as the interconnect material are coated as bars on top of the electrolyte sheet.

This design has the following advantages:

i) no critical sealing request, a very simple system;

ii) very integrated, resulting in more economical construction and higher power density;

iii) the geometry shown in Fig. 6 takes advantage of the nanostructured thin film, since prior orientation of the thin film grains creates much higher ionic transport ability along the thin film plane than in the z direction, as discovered in Li_2SO_4–Al_2O_3, and –SiO_2 thin films [14];

iv) i) to iii) provide a highly cost effective system and simple manufacturing.

A concrete example for the stack construction is described as follows:

i) preparation of porous or dense Al_2O_3 supporting sheet by tape casting, 0.5 to 1.0 μm thick, size larger than 100 cm^2; pre-sintering;

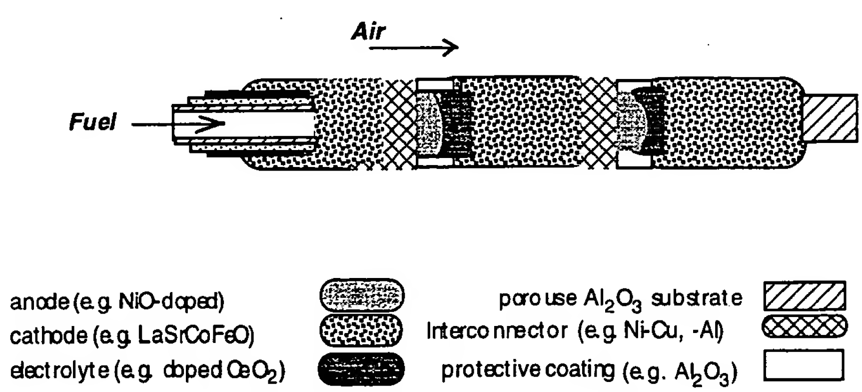

Fig. 5. Single tubular integrated fuel cell device/stack.

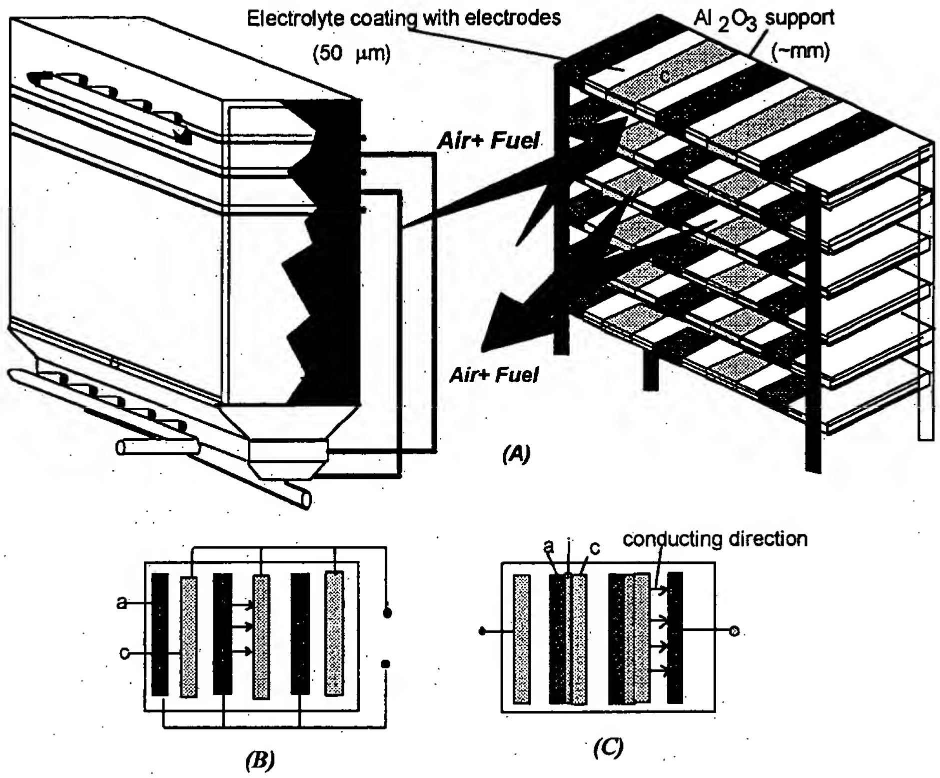

Fig. 6. New intermediate temperature FC planar stack design (A). (B) and (C) show a top view of a plate in different configurations, a: anode; c: cathode; i: interconnect.

ii) preparation of the electrolyte membranes by tape-casting, about 50 μm thick, on both sides of the Al_2O_3 support;
iii) sintering of the electrolyte membranes on the support;
iv) using the sol–gel technique to prepare thin film electrolyte layers, about 10 μm thick, on both sides of the sintered sheet to smooth the electrolyte surface. The smoothed surface is very important for achieving a high performance of the FC device;
v) application of several tens to 100 μm thick electrodes (anode and cathode) using gas deposition, sputtering or screen printing processes, and completion of the construction of the complete FC stack.

The above design and fabrication of intermediate temperature FC plants are based on new innovations for developing a new generation of advanced intermediate temperature FC technology. It has, however, a long way to go until it reaches the stage of practical devices/plants.

4. Remarks

The development of materials, technology and systems may have to take new directions in order to develop new FC generations. Although the studied FCs have a long way to go until they are practical power sources, they indicate the possibility of developing applications with importance for science and technology.

Acknowledgements

This research is supported on the Swedish side by: TFR (the Swedish Research Council for Engineering Sciences) and STINT (the Swedish Foundation for International Co-operation and Higher Education); on the Chinese side by: NSFC (National Nature Science Foundation of China) and MSTC (Ministry of Science and Technology of China).

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